

component consisting of a sulfur-lean, mono-aromatic-rich fraction to obtain components for refinery blending of transportation fuels.

Kindly amend Claim 12 to read as follows:

12. The process according to claim 21 wherein the immiscible
5 peracid-containing phase is an aqueous liquid formed by admixing,
water, a source of acetic acid, and a source of hydrogen peroxide in
amounts which provide at least one mole acetic acid for each mole of
hydrogen peroxide.

REMARKS

10 This amendment is respectfully submitted to place subject Application in condition for allowance. Claims 1 to 11, inclusive, and Claim 13 have been canceled without prejudice. Claims 21 to 25 have been entered to more distinctly claim the subject matter of the invention, and Claim 12 has been amended to depend from
15 Claim 21.

Claim 21 is directed to Applicants' novel process for the production of refinery transportation fuel or blending components for refinery transportation fuel, which process comprises: (a) hydrotreating a petroleum distillate consisting essentially of
20 material boiling between about 50° C. and about 425° C. by a process which includes reacting the petroleum distillate with a source of hydrogen at hydrogenation conditions in the presence of a hydrogenation catalyst to assist by hydrogenation removal of sulfur and/or nitrogen from the hydrotreated petroleum distillate; (b)
25 fractionating the hydrotreated petroleum distillate by distillation to provide at least one low-boiling blending component consisting of a sulfur-lean, mono-aromatic-rich fraction, and a high-boiling feedstock consisting of a sulfur-rich, mono-aromatic-lean fraction;
30 (c) contacting at least a portion of the high-boiling feedstock with an immiscible aqueous phase comprising at least one organic peracid or precursors of organic peracid, in a liquid reaction

mixture maintained substantially free of catalytic active metals and/or active metal-containing compounds and under conditions suitable for oxidation of one or more of the sulfur-containing and/or nitrogen-containing organic compounds including
5 temperatures in a range upward from about 50° C. to about 150° C; (d) separating at least a portion of the immiscible peracid-containing phase from the reaction mixture to recover an essentially organic phase from the reaction mixture; and (e)
10 treating at least a portion of the recovered organic phase with a solid sorbent, an ion exchange resin, and/or a suitable immiscible liquid containing a solvent or a soluble basic chemical compound, to obtain a product containing less sulfur and/or less nitrogen than the feedstock. Support for this claim is found in the Specification, for example, at page 18, lines 20 to 23, and now canceled Claims 1
15 to 11.

Claim 22 recites Applicants' novel process according to claim 21 wherein the conditions of oxidation include temperatures in a range upward from about 80° C. to about 125° C. and sufficient pressure to maintain the reaction mixture substantially in a liquid phase. Support for this claim is found in the Specification, for example, at page 18, lines 20 to 23.

Claim 23 recites Applicants' novel process according to Claim 21 which further comprises treating at least a portion of the separated peracid-containing phase to remove and at least a portion of the sulfur-containing and nitrogen-containing organic compounds and water contained therein, and thereafter recycling to the reaction mixture at least a portion of the treated peracid-containing phase having a water content of less than 60 percent by volume. Support for this claim is found in the Specification and, for example, in now canceled Claims 1, 4 and 13.
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Claim 24 recites Applicants' novel process according to claim 21 wherein the high-boiling oxidation feedstock consists essentially

of material boiling between about 200° C. and about 425° C. Support for this claim is found in the Specification and, for example, in now canceled Claim 9.

Claim 25 recites Applicants' novel process according to claim 21
5 further comprising blending the product containing less sulfur and/or
less nitrogen than the oxidation feedstock with at least a portion of the
blending component consisting of a sulfur-lean, mono-aromatic-rich
fraction to obtain components for refinery blending of transportation
fuels. Support for this claim is found in the Specification and, for
10 example, in now canceled Claim 8.

In view of the amendments submitted herein and previous
filed papers, it is the position of Applicants that the instant
Application is in condition for allowance.

Applicants and their undersigned Attorney note with
15 appreciation that Examiner Griffin has withdrawn rejection as
described in paper no. 6 in view of amendments filed on February
10, 2003 with a declaration under 37 CFR 1.131 to overcome the
Rappas reference of record.

Claim Rejections - 35 U.S.C. § 103

20 In the outstanding Office Action, Claims 1 to 13 and 15 to 18
were rejected under 35 U.S.C. § 103(a), as being unpatentable over
U.S. Patent No. 1,972,102 in the name of William M. Malisoff
(Malisoff), in view of U.S. Patent No. 6,217,748 in the name of
Shigeto Hatanaka, Osamu Sadakane, and Hideshi Iki and assigned to
25 Nippon Mitsubishi Oil Corp. (JP), (Hatanaka et al.), and U.S. Patent
No. 6,274,785 in the name of Walter Gore (Gore).

The Malisoff reference of record describes a treatment for
removal of sulfur from hydrocarbon oil by treating a hydrocarbon
oil with a **miscible** treating solution containing a concentrated
30 water solution of hydrogen peroxide and sufficient quantity of

acetic acid to render the treating solution **miscible** with the oil, and thereafter adding an equivalent volume of water to obtain a resulting dilute aqueous phase which separates from the treated oil phase (Malisoff, for example, at page 1, lines 49 to 59, and page 2, lines 3 to 5). In example 1 of Malisoff, the resulting dilute aqueous phase containing more than 60 percent water (calculated without including any water formed by reaction of hydrogen peroxide).

By contrast, instant claims include a key step of contacting a high-boiling, sulfur-rich, mono-aromatic-lean, petroleum distillate fraction with an **immiscible (non-miscible)** phase comprising at least one organic peracid or precursors of organic peracid in a liquid phase reaction mixture. Maintaining the reaction mixture substantially free of catalytic active metals and/or active metal-containing compounds and under conditions suitable for oxidation of one or more of the sulfur-containing and/or nitrogen-containing organic compounds including temperatures in a range upward from about 50° C. to about 150° C. are recited elements of the invention. Blending components containing less sulfur and/or less nitrogen than the oxidation feedstock are recovered from the reaction mixture.

Advantageously, at least a portion of the immiscible peracid-containing phase is also recovered from the reaction mixture and recycled to the oxidation. Integrated processes of this invention may also provide their own source of high-boiling oxidation feedstock derived from other refinery units, for example, by hydrotreating a petroleum distillate.

It is the position of Applicants that their novel processes differ in kind, not merely in degree, from the process described in the Malisoff reference of record.

Oils treated according to examples in the Malisoff reference of record initially contained sulfur at levels of 0.21, 1.12, and 0.25 percent sulfur. However the products of the Malisoff treatments, resulted in products containing sulfur levels of 0.16, 0.88 and 0.10

percent sulfur (1,600 ppmw, 8,800 ppmw and 1,000 ppmw). These levels of sulfur are very high in view of sulfur specifications for transportation fuels. Legislation on sulfur in diesel fuel in Europe, Japan and the U.S. has recently lowered the specification to 0.05
5 percent by weight maximum sulfur in Diesel fuel, and indications are that future specifications may go far below the current 0.05 percent by weight level.

By contrast, all products of an alumina treatment according to Applicants' Example 14 had a sulfur concentration of less than 3
10 ppmw, and in general 1 ppmw sulfur.

By the contacting step that instant claim recite, the selected petroleum distillate fraction with a peracid-containing immiscible phase in a liquid reaction mixture maintained substantially free of catalytic active metals and/or active metal-containing compounds, the
15 tightly substituted sulfides are oxidized into their corresponding sulfoxides and sulfones with negligible if any co-oxidation of mononuclear aromatics. These oxidation products due to their high polarity, can be readily removed by separation techniques such as sorption, extraction and/or distillation. The high selectivity of the
20 oxidants, coupled with the small amount of tightly substituted sulfides in hydrotreated streams, makes the instant invention a particularly effective deep desulfurization means with minimum yield loss. The yield loss corresponds to the amount of tightly substituted sulfides oxidized. Since the amount of tightly substituted sulfides present in a
25 hydrotreated crude is rather small, the yield loss is correspondingly small.

It is the position of Applicants that the Malisoff reference of record does not suggest the essence of their novel multi-step process as recited according to instant Claims 12 and 14 to 25
30 inclusive.

The Hatanaka et al. reference of record describes a four-step hydrodesulfurization (HDS) of a sulfur-containing petroleum hydrocarbon diesel gas oil. The gas oil is catalytically hydrodesulfurized at 300° C. to about 500 ppm sulfur in a first HDS step. Step two is distillation at a cut point temperature in a range of 320 to 340° C. Step three is a second HDS of only the heavy fraction of the distillation at up to 380° C. The twice hydrodesulfurized heavy fraction (90 ppm sulfur) is mixed with the light fraction of the distillation (10 ppm sulfur) to a sulfur content of up to 0.01%. For example, a product having 50 ppm sulfur is reported by Hatanaka et al. as Example 3.

As noted above, all products of an alumina treatment according to Applicants' Example 14 had a sulfur concentration of less than 3 ppmw, and in general 1 ppmw sulfur.

Obvious disadvantages of the Hatanaka et al. process include the costs of the additional hydrogen and energy required for both the distillation and further hydrodesulfurization steps. While Hatanaka et al. state the hydrogen sulfides from the two HDS steps may be removed from the off-gas by an amine absorption arrangement, carbon oxides are also by-products of the HDS steps. Generation of the additional hydrogen also produces more carbon dioxide. These are especially critical at a time when most industries are striving to limit the amount of emitted "green house" gas such as carbon dioxide.

Less obvious disadvantages of the second HDS in the Hatanaka et al. process include a loss of non-sulfur multi-ring aromatic hydrocarbons to saturated non-aromatic ring compounds. This has negative fuel property/quality consequences such as affecting the pour point, reducing the lubricity, and reducing the fuel value (Btu/volume) of diesel fuel. A diesel fuel rich in aromatics have a higher Btu/volume and give more miles per gallon.

The oxidation step in Applicants' novel process is very specific in the targeting of sulfur heteroaromatic compounds. Oxidation of these offending sulfur heteroaromatics is selective and other aromatic hydrocarbons not containing sulfur or nitrogen are 5 unaffected by the process.

Danping Wei and H. A. Spikes reported results of their investigation of the main chemical factors influencing the lubricity of diesel fuels, diesel fuel fractions and model fuels in a paper titled THE LUBRICITY OF DIESEL FUELS (Wear, 111 (1986) 217 - 235).
10 Under test conditions the used, sulfur-containing components were found to be pro-wear. Polycyclic aromatic species and, in particular, oxygen-containing components, were found to reduce wear significantly.

Contrary to the position of Examiner, substituting the 15 oxidation treatment of Malisoff for the second HDS of Hatanaka et al. does not produce an equivalent result, because as shown above, the oxidation treatment of Malisoff is different in kind from instant claims which include a key step of contacting a high-boiling, sulfur-rich, mono-aromatic-lean, petroleum distillate fraction with an 20 **immiscible (non-miscible)** phase comprising at least one organic peracid or precursors of organic peracid in a liquid phase reaction mixture.

By contrast Applicants' novel process includes a catalyzed, hydrogen peroxide, promoted oxidative desulfurization of a sulfur-rich, mono-aromatic-lean fraction without further production of 25 carbon dioxide or hydrogen sulfides. An additional advantage of Applicants' novel oxidative desulfurization process is that oxidation is selective to sulfur and nitrogen containing heteroaromatic compounds, i.e. does not saturate the non-sulfur, non-nitrogen 30 multi-ring aromatic hydrocarbons.

Examiner used the Gore reference of record to disclose treating and recycling the oxidant. However, practice of recycle according to the disclosure of Gore on hydrogen peroxide and acetic acid results in a critical accumulation of a high water concentration.

- 5 Each mole of hydrogen peroxide used results in formation of mole of water. As water concentration increases, the equilibrium concentration of peracetic acid is reduced, which must decrease the efficiency of the oxidation.

By contrast, Claim 23 recites Applicants' novel process according
10 to claim 21 further comprising treating at least a portion of the separated peracid-containing phase to remove and at least a portion of the sulfur-containing and nitrogen-containing organic compounds and water contained therein, and thereafter recycling to the reaction mixture at least a portion of the treated peracid-containing phase
15 having a water content of less than 60 percent by volume.

Examiner used the GB reference of record to disclose treatment of a treated oil with an alumina adsorbent. However, according to the GB reference an oxidized hydrocarbon material is washed with 2 Normal sodium sulfite and thereafter washed with water to remove
20 residual sodium sulfite, prior to treating the twice washed hydrocarbon material with the alumina adsorbent.

Claims 19 and 20 were rejected under 35 U.S.C. § 103(a), as being unpatentable over U.S. Patent No. 1,972,102 (Malisoff), in view of U.S. Patent No 6,217,748 (Hatanaka et al.) and U.S. Patent
25 No. 6,274,785 in the name of Walter Gore (Gore) as applied to Claim 12, and further in view of U.S. Patent No. 3,163,593 in the name of Alfred Bentley Webster, Norman James Herbert, and Richard Rigby (Webster et al.). Applicants respectfully traverse these rejections.

Examiner used the Webster et al. reference of record to
30 disclose treatment of an oil obtained from an oxidizing process with

an alkaline material as suggested by Webster because a material with a reduced amount of sulfur will be obtained.

Contrary to the position of Examiner, Webster et al. report their thermal treatment in the presence of lime of an oil obtained from an oxidizing process, resulted in a material having a higher level of sulfur than treatment without lime, 2.52 percent sulfur with lime, 2.03 percent sulfur without lime (Webster et al. column 4, lines 25 to 45).

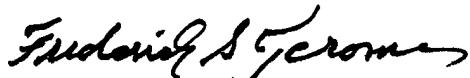
It is the position of Applicants that instant Claims 12 and 14 to 25 inclusive, meet all requirements under 35 U.S.C. § 103(a).

Base on the amendments submitted herein, and previously, and Paper No. 8 declaration under 37 CFR § 1.131, Applicants urge that Claims 12 and 14 to 25 inclusive, all claims now presented, are in condition for allowance. Applicants respectfully request Primary Examiner Griffin to pass subject application for allowance.

Do not hesitate to contact Frederick S. Jerome whose telephone number is (630) 832-7974 (FAX (630) 832-7976) if additional assistance is needed regarding this paper or earlier papers for Applicants.

Applicants and their undersigned Attorney appreciate Examiner's attention and further consideration of this matter.

Respectfully submitted,



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Version of Amended Claims with Markings
to Show Changes Made

12. (Once amended) The process according to claim 21
5 [11] wherein the immiscible peracid-containing phase is an aqueous liquid formed by admixing, water, a source of acetic acid, and a source of hydrogen peroxide in amounts which provide at least one mole acetic acid for each mole of hydrogen peroxide.